Synthetic Fuels from Coal-Heavy Oil Mixtures

Phase 1 Report

"A Survey of Work Applicable to Coal Bitumen Mixtures as Synthetic Fuels Feedstocks"

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MICROMEDIA

Introduction

A survey has been made for significant research and development being performed worldwide that is applicable to the co-conversion of coal and bitumen to synthetic liquid fuels. Table 1 (attached) summarizes the contacts which were made to acquire information. The following discussion summarizes our findings.

Historical-Coal

Literature on the liquefaction of coal extends backward for over 60 years. Many attempts have been made (1) (and are still being made) to understand the composition of coal and its reaction with various "solvents" under various conditions. A commercial coal hydrogenation industry evolved in Germany in the 1930s (2). The installations were heavily damaged during World War II, and the ready availability of crude oil thereafter discouraged continued operation.

During the past 15-20 years, renewed attempts have been made to solvate or liquify coal (3). An extensive pilot plant demonstration program has been in progress in the U.S. to develop a process to convert coal into clean liquid fuel, i.e. into a "synthetic" crude oil (4). A more modest effort has been in progress in West Germany, and lesser efforts in Poland, Australia, Japan, Great Britain, and South Africa.

The major processes which have been piloted in the U.S. are the Solvent Refined Coal (SRC) process, the H-Coal process, and the Exxon Donor Solvent (EDS) process. These processes have in common the feature that coal, slurried with a recycled process solvent, is subjected to elevated temperature and pressure in an atmosphere which is rich in hydrogen. The processes differ in the manner of application of catalysts. In the H-Coal process, the reactions with coal are carried out in the presence of a catalyst in an expanded (ebullated) bed. In the EDS process, the recycle oil is catalytically hydrogenated before contacting the coal and no catalyst contacts the coal. In the SRC process, no catalyst is applied in the primary liquefaction step; the process relies on the properties of the coal and its ash to accelerate the reactions.

These processes all produce primary oils which are much more aromatic than natural crude oil fractions and which contain higher concentrations of oxygenated hydrocarbons. Hence, they require subsequent severe hydrogenation to prepare synthetic crude oils which are suitable for existing crude oil refineries.

Bitumen

Bitumen from the Athabasca oil sands is currently being converted to a "synthetic" crude oil by thermal coking followed by hydrogenation of the virgin and coker distillate fractions. A highly refined (low sulfur, low nitrogen, no residue) distillate oil is produced which is a premium feedstock for existing crude oil refineries.

With this as background, we are looking for a process combination which combines the processing of coal and bitumen and is aimed toward the production of a synthetic crude oil. We are likely looking for a process in

which the bitumen serves as the slurry medium for coal, thereby increasing the net feed capacity of the process. We are looking for any synergism which improves process efficiency, liquid yields, or product selectivity over separate processing of coal and bitumen.

Canada

Perhaps not surprisingly, the major effort which we have located is that of the Alberta Research Council (5-11). They have performed batch autoclave experiments in which bituminous and subbituminous coals were contacted with Athabasca and Cold Lake bitumens, Lloydminster heavy crude oil and distillates derived from Athabasca bitumen. The contacting was done with and without hydrogen pressure, and with and without catalysts. The effects of residence time, pressure and reaction temperatures were surveyed. Promising results were obtained in some instances, i.e., coal solubilization was equivalent to that obtained with tetralin, which is often used as a model compound in donor solubilization studies.

It is pointed out that as an organic solvent, for example, a petroleum fraction, acts as a hydrogen donor, it is degraded to a lower H/C ratio, which must be restored by adding hydrogen. This may be done in-situ, as in the H-Coal process or in an external reactor as in the EDS process. We note that Gulf Canada, Ltd. (12) has developed the Donor Refined Bitumen (DRB) process to improve product selectivity, i.e. reduce residuum production, in the refining of Athabasca bitumen. Coal could be injected into this process via the donor recycle stream as well as with the bitumen stream.

United States

Hydrocarbon Research Inc. (HRI), a subsidiary of Dynalectron Corporation, offers the COIL™ process (13), a companion to their H-Oil and H-Coal processes, for the coprocessing of coal with a petroleum residuum fraction. A summary of data from processing 1/0, 1/1, 1/2 and 0/1 W/W coal/residuum mixtures shows apparent synergistic effects on distillate oil yield, residual oil yield, and fuel oil desulfurization in the runs in which coal was admixed with residuum. A laboratory program, such as that being pursued by the Alberta Research Council, is suggested to arrive at process parameters and yields for given coals and bitumen fractions. Properties of the products must be determined and process requirements to upgrade them to marketable commodities must be established.

Exxon indicated that they have done no work with crude oil fractions in the EDS coal liquefaction program. However, they have done some proprietary work and have a patent (USP 4,298,454) which covers the use of crude oil fractions for coal/oil slurry feeds to liquefaction.

UOP has no work in progress on coal/oil slurries but have two patents (USP 3,705,092 (1972) and USP 4,338,183 (1982) which covers the use of heavy oils as "solvents" in a coal hydroliquefaction process.

Chevron reports that they have done some work on the co-processing of coal with crude oil fractions but have not as yet released it.

Cities Service is co-funding, with DOE and Auburn University, a two year $290,000 program at Auburn University on the evaluation of oil refinery streams as feedstocks with coal in a coal hydroliquefaction process (see Coal R&D, November 3, 1982). The program will include a laboratory study of reaction temperature, pressure, catalysis, solvent composition and solvent/coal ratio. The program is an outgrowth of Cities Service's involvement in the SRC-I process, in two-stage coal liquefaction and in the LC-Fining process. It is aimed at exploring how coal liquefaction might be advantageously integrated into an oil refinery.

Gulf has done some work on the liquefaction of coal with oil which they do not wish to disclose. Sohio reported that they had done no work on coal/oil mixtures.

Europe

Veba Oel AG reported at a recent EPRI conference (14) that the Federal Republic of Germany had requested them to perform a study for a commercial sized plant to hydrogenate about 4.3 million ton per year of raw coal, or alternatively 2.1 million ton per year of raw coal plus 5.5 million ton per year of heavy oil. Yield data were reported only for the coal case; when contacted for additional information, Veba indicated that they had done no experimental work on co-processing of coal and petroleum fractions.

Nynas Petroleum in Sweden is known to be interested in both coal conversion and in Venezuelan tar sands. However, when contacted, Nynas replied that no experimental work had been done there on coal/oil mixtures.

Japan

The Japanese have been pursuing an extensive program on coal liquefaction--studying the effects of solvents, catalysts, gas atmosphere, etc. on the conversion of coal to liquids. Data were reported at the just completed Pan Pacific Synfuels Conference (15). No data were given that are specific to coal/oil processing. Private discussions with Mitsubishi Heavy Industries (MHI) revealed that they had evaluated coprocessing of coal with petroleum oils. It appears that coprocessing did not offer sufficient incentive over separate processing to justify continuation of the program. For example MHI no longer uses a petroleum derived recycle solvent in the pilot-scale coal liquefaction (solvolysis) work. Asahi Chemical has conducted batch autoclave hydro-liquefaction tests on the coal/oil mixtures. We have no data on this work.

The Japanese (Osaka Gas Co., Ltd) offer the Cherry-P process (16), in which a small amount of coal is added to a vacuum residue to suppress coke deposition on reactor surfaces during thermal cracking. You have, of course, followed the same procedure in the CANMET hydrocracking process (16, 17).

Ranking of Applicable Research

Synthetic Fuels Associates has ranked the importance of the various research programs applicable to coprocessing coal-bitumen mixtures

(Table 2). The most extensive and significant effort which we have located appears to be that in progress by the Alberta Research Council, followed by that done by HRI. The Cities Service/Auburn University program could develop significant results over the next two years. UOP has been issued two patents (not reviewed) which indicate that they may have done significant work. There is no information upon which to judge the significance and extent of the work done by those shown in Tables 1 and 2 as having done proprietary work, i.e., Exxon, Gulf, Chevron, Mitsubishi, and Asahi.

Conclusions and Recommendations

We have identified several proprietary research programs and have found a limited amount of data on the hydro-liquefaction of coal/heavy fractions. Major current efforts which we have located are the Alberta Research Council and the Auburn University programs. We are prepared to proceed to Phase 2 and prepare a "Technical Analysis" of the project from the data which we have identified.

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Table 1

ORGANIZATIONS CONTACTED AND PERTINENT COMMENTS

No.	Organization	Comments
1.	HRI/Dynalectron Corp.	COIL™ process - brochure available
2.	Exxon	Proprietary work U.S. Patent 4,298,454
3.	Chevron	Proprietary work - still confidential
4.	UOP	No work in progress now, U.S. Patents 4,338,183 (1982) and 3,705,092 (1972)
5.	Gulf	Proprietary work but do not wish to disclose
6.	Sohio	No work done
7.	Cities Service	Have considered integration into an oil refinery. Now co-sponsoring work with DOE and Auburn University
8.	Veba Oel	Have considered in evaluation studies only; no experimental work
9.	Nynas	No applicable work
10.	Mitsubishi	Have done some work; now discontinued
11.	Asahi Chemical	Have done some autoclave processing

Table 2

RANKING OF IMPORTANCE OF RESEARCH WORK
APPLICABLE TO COAL-HEAVY OIL MIXTURES

Rank	Organization	Comments
1.	Alberta Research Council	Extensive research with coal/ bitumen mixtures
2.	Hydrocarbon Research Inc.	COIL™ process
3.	Cities Service/Auburn University	Research program underway coprocessing coal/oil mixtures
4.	UOP Inc.	Two expired patents on hydroliquefaction of coal/oil slurries
No basis for ranking	Asahi Chemical, Chevron, Exxon, Gulf, MHI	Proprietary research performed